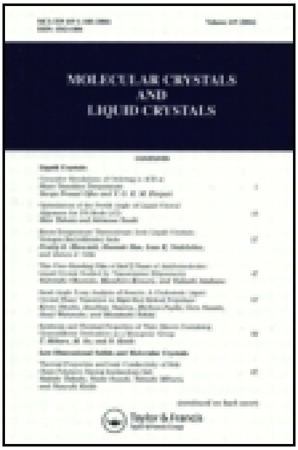

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"Shear Acoustic Wind" Acting on Particles Embedded in a Liquid Crystal

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We present here a theory of a specific force acting on the particles embedded in a liquid crystal (LC) if a "fast" (viscous) shear wave is excited in the LC layer. The origin of this force is very similar to the so-called "ponderomotive forces" acting on the electrons or bubbles in liquids in the presence of spatially non-homogeneous periodic fields (electric in the first case and acoustic in the second). We show that one can use this effect for manipulation, orientation through sedimentation, and separation of particles, including carbon nanotubes. This article is mostly focused on the case of elongated particles in a nematic liquid crystal medium. However, more general consideration is also presented.

1. INTRODUCTION

The advancement of nano-technology and, in particular, the development of new useful materials and devices require post-synthesis separation of nano-particles with different properties and controlled assembly of nano-scale building blocks such as single-wall (SWCNT) and multi-wall (MWCNT) carbon nanotubes (CNT), which are the most promising blocks for many applications. Post-synthesis strategies of organizing nanoparticles include self-assembly directed by specific interparticle interactions, templating by patterned substrate or on the surface of larger particles, mechanical ordering, and use of external fields [1].

Application of liquid crystals (LC) for controlled manipulation and assembling of nano-sized objects is considered a promising approach due to the unique properties of liquid crystalline fluids (hereafter referred to as LC), The manipulation of the particles embedded in a

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liquid crystal is now regarded as a possible way to assemble arrays of nano-tubes, separate particles, etc. [1].

The application of liquid crystals as "smart solvents" is based on their unique properties. LCs represent an intermediate state between regular liquids and crystals. Still liquid in terms of the mobility of their molecules, LCs manifest long-range orientation (nematics) and spatial (smectics) order [2]. Foreign particles, embedded in an LC medium, distort LC molecular fields in their neighborhood and change the overall energy of the system. The energy of the whole system, which includes the LC and particles, depends on the chemical properties and symmetry of the LC, size, shape, orientation in respect to the LC director and chemical properties of the embedded particle and on the presence of an external field. A system tends to come to the state with minimal overall energy. As a result, appropriate generalized forces acting on the particles embedded in the LC appear. Specific "liquid crystalline forces" tend to orient asymmetric and/or chemically non-homogeneous particles with respect to LC axes of symmetry and push particles in the specific spatial positions to provide minimum energy, if the molecular field of LC is non-homogeneous, etc.

The ability of LCs to provide forces on embedded particles open new opportunities. The orientation effect of LCs on elongated particles was understood very early on in the history of liquid crystal research. Brouchard and de Gennes in their pioneering article [3] proposed to use solvents in the LC state for the creation of liquid ferromagnets (ferronematics) by the introduction of elongated magnetic particles in nematic LC. Relatively recently, M. D. Lynch and D. L. Patrick used LCs to achieve oriented sedimentation of CNTs [1]. S. Lopatnikov and V. Namiot in 1978 invented and described specific long-range interactions between particles embedded in liquid crystals [4]. The physics of this interaction can be explained as follows: each particle distorts the LC molecular field on its neighborhood. If the distortions by different particles LC regions overlap, the energy of the system will depend not only on the shape and orientation of the particles but also on their relative positions. As a result, an interaction force between particles appears. S. Lopatnikov and V. Namiot calculated this force for particles of arbitrary shape embedded in nematic and smectic liquid crystals in both the absence and presence of the magnetic fields. The researchers showed that this interaction is sensitive to the shape of particles and presence of external, specifically, magnetic fields. Namiot and Lopatnikov pointed out that the advantage of using LCs as "smart solvents" for colloids with complex shapes is that a "key-lock interaction" takes place during the interaction of particles [4,5]. The major theoretical prediction of this article for the particular case of small droplets found experimental proof in 1997 [6]. On the basis of a similar general idea, Namiot and Lopatnikov proposed using the effect of the dependence of the elastic module of LCs on temperature to speed up the process of sedimentation and separation of macromolecules. In accordance with their work, a gradient of temperature provides a gradient of the "chemical potential" of the particle embedded in an LC. The appropriate "chemical potential" includes the surface energy of an LC–particle interaction and energy of LC distortion. The gradient of the "chemical potential" provided by the temperature gradient leads to the appearance of the force acting on the particle and pushing it in the region with higher temperature (or "out," depending on the chemical properties of the particles) [5].

In spite of this progress, developing new methods for the manipulation of particles embedded in a liquid crystal medium is still of great interest. In this article, we consider a new physical effect, which can be used for this purpose. In contrast to the above-mentioned forces, which appear if some static distortion of the LC molecular field exists, the nature of the considered effect is related to the gradient of the time average increase of the energy of a distorted by the embedded particle region of LC in a spatially non-homogeneous periodic perturbation of the LC molecular field.

To explain qualitatively the physical nature of the effect, let us consider a volume of nematic liquid crystal homogenously oriented in space. The embedded particle will distort the LC director field. In a general case, the energy associated with the distortion of the LC caused by the presence of the particle will depend on the relative orientation of the particle with respect to the orientation of the LC director far away (Fig. 1). In equilibrium, to minimize the distortion energy, the particle will obtain some preferable orientation with respect to the far-field orientation of the LC director. Consequently, the inclination of the particle from this preferable orientation will cause an increase in the distortion energy of the LC.

Let us suppose that one can provide by some means, for example, by generation of a director wave, periodic inclination of a particle from its preferable orientation. In this case, the average energy of the distorted region in the presence of the periodic perturbation will be higher than its energy in the absence of the perturbation. In a general case, this average force $\langle \mathbf{f} \rangle$ acting on the embedded particle in a non-homogeneously perturbed periodic director field will be equal to

$$\langle \mathbf{f} \rangle(\mathbf{X}) = -\nabla_x \langle E(\mathbf{X}\Phi(\tau)) \rangle. \tag{1}$$

Here, $\mathbf{X}$ is the coordinate of the particle's center of mass; $\hat{\Phi}$ is a value, characterizing the inclination of the particle from its preferable orientation with respect to the far-field orientation of the LC director; $E(\mathbf{X}\hat{\Phi}(t), \mathbf{n}(\mathbf{x}_S, t))$ is the energy of the periodically distorted region.

Thus, if the intensity of the perturbation is spatially nonhomogeneous, the average gradient of energy of the distortion energy will not be equal to zero, and the associated average force pushing the particle out from the region of higher intensity of perturbation will appear.

It is clear from the above qualitative discussion that the described “first order” effect cannot be observed for spherical particles due to their symmetry, or for “lightweight” elongated particles that are “frozen” in an LC so that they always stay practically parallel to the local average director field. In this case, the second-order effect must be taken into account.

The physical nature of the second-order effect is slightly different from the nature of the first-order effect. The second-order effect’s appearance is related to periodic changes of the curvature of the director field.

The difference between first- and second-order effects is that, in the first case, this addition of energy is related to the inertia of the particle, which cannot immediately follow the change in the time orientation of the LC director, and in the last case, the particle follows the director only “in average.” However, if the size of the particle is

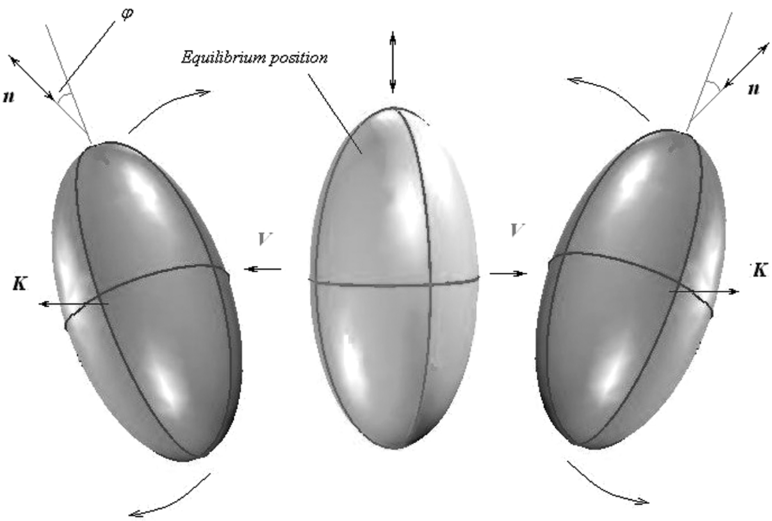

FIGURE 1 On the physical nature of the first-order acoustic shear wind effect.

comparable with the characteristic length of director perturbation, the local director is not collinear to the particle (Fig. 2), and thus, some addition to the energy appears. We will consider the second-order effect when applied to a force acting on the long rods. However, it still works for the spherical particle, too, by the physical nature of this effect related with spatial non-homogeneities of the director field on the scale of the embedded particle.

Taking into account that the second-order effect is more universal and works for most common types of nano-particles, such as CNT, in this article we will focus just on this effect and will consider as an example the force acting in a non-homogeneous periodic field on an elongated rigid particle. We will consider the first-order effect elsewhere.

The effective way to provide controlled periodic perturbation of an LC molecular field and, in the meantime, a high gradient of average energy is to generate in LC the so-called "fast shear wave" that propagates in the liquid crystalline (nematic) medium [7]. This wave is an analogue of the regular viscous waves in regular viscous liquids.

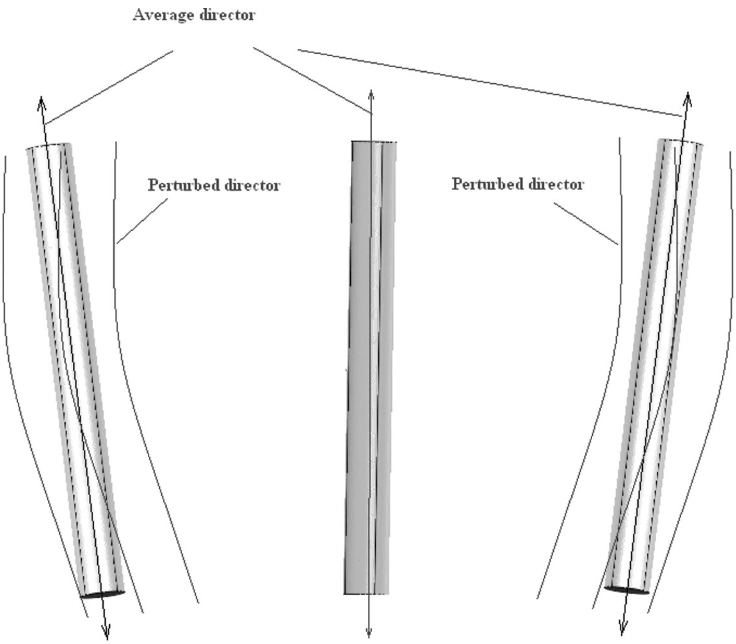

FIGURE 2 Origin of the second-order ponderomotive force acting on the particles embedded in nematic LC.

Specific to the "fast wave" in a liquid crystal is that in this wave the director of the LC is practically "frozen" in local liquid motion. In the meantime, the fast wave can be easily generated, for example, by the vibration of one wall of the liquid crystalline layer. The additional energy of the particle embedded in the LC related to the presence of the shear viscous wave is proportional to its intensity, which decreases exponentially with the distance from the vibrating wall. As a result, the force trying to push the particle out of the region of higher intensity of the shear wave will appear. In spite of the generation of shear viscous wave is not the only way to reach the goal, due to the relative simplicity of this way to reach the goal, we will refer the effect related with inclination of particle orientation from orientation of the LC director as "*shear acoustic wind*." We will show that this effect seems to be strong enough to compete with the hydrodynamic method of particle sedimentation.

2. THEORY OF THE FORCES ACTING ON THE ELONGATED FOREIGN PARTICLE IN NON-HOMOGENEOUS PERIODIC DIRECTOR FIELD

To make the physics of the effect clear, we will use a simplified assumption that the director field of the liquid crystal is "frozen" in the fluid. Let us consider a sufficiently long, ideally rigid paiticle embedded in a non-perturbed nematic LC having initial homogeneous orientation $\bar{\mathbf{n}}_0$ forming the tilt angle Θ with the limiting surface S (Fig. 3) and let us characterize the orientation of the particle by a unitary vector $\mathbf{N}$, directed along its long axis.

The energy of interaction of the particle with a liquid crystal is a function of $\bar{\mathbf{n}}_0$ and $\mathbf{N}$, satisfying some properties of symmetry.

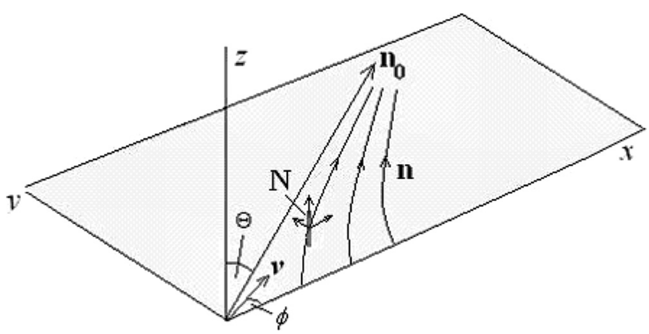

FIGURE 3 The geometry of the problem.

Specifically, the inversion of the direction of the director $\bar{\mathbf{n}}_0$ and/or the vector $\mathbf{N}$, as well as the arbitrary spatial rotation of the system as a whole must not change the energy of the system.

Let us suggest that a particle is oriented in this case along the LC director. This means that for small inclinations of the particle orientation from the LC director, the energy of particles can be presented as

$$E_t = E_{\max} - (E_{\max} - E_{\min})(\bar{\mathbf{n}}_0 \cdot \mathbf{N})^2. \tag{2}$$

This energy satisfies the requirements of the symmetry. It reaches the minimum if the vector $\mathbf{N}$ is parallel or (due to symmetry of LC molecules and particle) anti-parallel to director vector $\bar{\mathbf{n}}_0$ and the maximum, if the particle is oriented orthogonally to the director.

Let us now suggest that an acoustic wave distorts this state of the LC and provides a non-homogeneous state within the LC, which one can characterize by the non-homogeneous director field. The director field now becomes a function of coordinate and time: $\bar{\mathbf{n}} = \bar{\mathbf{n}}(\mathbf{x}, t)$.

Our major suggestion is that the long rigid particle interacts with the LC locally, and the energy of interaction of each "small piece" of the particle can be described locally by Eq. (5). The total energy of the particle can be, thus, presented as

$$E(\mathbf{X}, \mathbf{N}, t) = E_{\max} - (E_{\max} - E_{\min})\left\langle (\bar{\mathbf{n}} \cdot \mathbf{N})^2 \right\rangle_l. \tag{3}$$

We denote here

$$\langle \ldots \rangle_l = \frac{1}{L}\int_{-\frac{L}{2}}^{\frac{L}{2}} (\ldots)dl. \tag{4}$$

Let us present the director field as

$$\bar{\mathbf{n}} = \langle \bar{\mathbf{n}} \rangle_l + \tilde{\bar{\mathbf{n}}}. \tag{5}$$

Thus, by definition

$$\left\langle \tilde{\bar{\mathbf{n}}} \right\rangle_l = 0. \tag{6}$$

The energy of interaction can be presented now as

$$E(\mathbf{X}, \mathbf{N}, t) = E_{\max} - (E_{\max} - E_{\min})\left((\langle \bar{\mathbf{n}} \rangle_l \cdot \mathbf{N})^2 + \left\langle (\tilde{\bar{\mathbf{n}}} \cdot \mathbf{N})^2 \right\rangle_l \right). \tag{7}$$

The cross term in parentheses in (7) vanishes identically because of the property (6).

Let us next introduce the modulus of the value $\|\langle\bar{\mathbf{n}}\rangle\|$ and the angle φ between $\langle\bar{\mathbf{n}}\rangle$ and $\mathbf{N}$. One can rewrite the expression for the energy as

$$E(\mathbf{X},\mathbf{N},t) = E_{\max} - (E_{\max} - E_{\min})\left(\left\|\langle\bar{\mathbf{n}}\rangle_l\right\|^2 \cos^2\varphi + \left\langle(\tilde{\bar{\mathbf{n}}}\cdot\mathbf{N})^2\right\rangle_l\right). \quad (8)$$

Let us suggest first that one can neglect the last term in parentheses in (8), because in the weak acoustic field it has a higher order than the first term.

In this approximation, it is immediately apparent that sin $\varphi_{\min} = 0$, which means that in local equilibrium the particle is oriented parallel to the average (over the particle length) direction of the director.

Thus, one can approximately state

$$\mathbf{N} = \frac{\langle\bar{\mathbf{n}}\rangle}{\|\langle\bar{\mathbf{n}}\rangle\|}. \quad (9)$$

Here we took into account that $\mathbf{N}$ is the unitary vector (opposite to $\langle\bar{\mathbf{n}}\rangle$). Thus, one has an approximate expression for minimal energy

$$E(\mathbf{X},\mathbf{N}) = E_{\max} - (E_{\max} - E_{\min})\left\{\|\langle\bar{\mathbf{n}}\rangle\|^2 - \left\langle\left(\tilde{\bar{\mathbf{n}}}\cdot\frac{\langle\bar{\mathbf{n}}\rangle}{\|\langle\bar{\mathbf{n}}\rangle\|}\right)^2\right\rangle_l\right\}. \quad (10)$$

Let us now express the values of interest through the parameters of the acoustic field.

One has

$$\bar{\mathbf{n}} = \mathbf{n}_0 + \tilde{\mathbf{n}}; \quad \|\bar{\mathbf{n}}\| = 1. \quad (11)$$

The second condition defines the specific relationship between $\mathbf{n}_0$ and ñ. One has

$$\|\bar{\mathbf{n}}\|^2 = \mathbf{n}\cdot\mathbf{n} = (\mathbf{n}_0 + \tilde{\mathbf{n}}\cdot(\mathbf{n} + \tilde{\mathbf{n}}) = l + 2(\mathbf{n}_0\cdot\tilde{\mathbf{n}}) + \tilde{\mathbf{n}}^2 = 1. \quad (12)$$

Thus,

$$2(\mathbf{n}_0\cdot\tilde{\mathbf{n}}) + \tilde{\mathbf{n}}^2 = 0. \quad (13)$$

From (11) and (13), one also has

$$\langle\bar{\mathbf{n}}\rangle_l = \mathbf{n}_0 + \langle\tilde{\mathbf{n}}\rangle_l, \quad (14)$$

$$\mathbf{n} = \tilde{\mathbf{n}} - \langle\tilde{\mathbf{n}}\rangle_l, \quad (15)$$

and from (13)

$$2(\mathbf{n}_0 \cdot \langle \tilde{\mathbf{n}} \rangle_l) + \left\langle \tilde{\mathbf{n}}^2 \right\rangle_l = 0. \tag{16}$$

Let us next express the value $\|\langle \bar{\mathbf{n}} \rangle\|^2$ in terms of variations of the director field.

One has

$$\begin{aligned} \left\|\langle \bar{\mathbf{n}} \rangle_l\right\|^2 &= (\mathbf{n}_0 + \langle \tilde{\mathbf{n}} \rangle_l)(\mathbf{n}_0 + \langle \tilde{\mathbf{n}} \rangle_l) = 1 + 2(\mathbf{n}_0 \cdot \langle \tilde{\mathbf{n}} \rangle_l) + (\langle \tilde{\mathbf{n}} \rangle_t)^2 \\ &= 1 + \left[2(\mathbf{n}_0 \cdot \langle \tilde{\mathbf{n}} \rangle_l) + \left\langle \tilde{\mathbf{n}}^2 \right\rangle_l\right] + (\langle \tilde{\mathbf{n}} \rangle_t)^2 - \left\langle \tilde{\mathbf{n}}^2 \right\rangle_l. \end{aligned} \tag{17}$$

However, in accordance with (16), the term in parentheses is identically equal to zero. Thus, one has

$$\left\|\langle \bar{\mathbf{n}} \rangle_l\right\|^2 \equiv 1 - \left(\left\langle \tilde{\mathbf{n}}^2 \right\rangle_l - \langle \tilde{\mathbf{n}} \rangle_l^2\right). \tag{18}$$

The second term in the parentheses in the expression for energy (10) is given by

$$\begin{aligned} \left\langle \left(\tilde{\bar{\mathbf{n}}} \cdot \frac{\langle \bar{\mathbf{n}} \rangle}{\|\langle \bar{\mathbf{n}} \rangle\|}\right)^2 \right\rangle_l &= \frac{\left\langle (\tilde{\mathbf{n}} - \langle \tilde{\mathbf{n}} \rangle_l)(\tilde{\mathbf{n}}_0 + \langle \tilde{\mathbf{n}} \rangle_l)^2 \right\rangle_l}{1 - \left(\left\langle \tilde{\mathbf{n}}^2 \right\rangle_l - \langle \tilde{\mathbf{n}} \rangle_l^2\right)} \\ &= \frac{\left\langle (\tilde{\mathbf{n}}\mathbf{n}_0 - \langle \tilde{\mathbf{n}} \rangle_l \mathbf{n}_0 + \tilde{\mathbf{n}}\langle \tilde{\mathbf{n}} \rangle_l - \langle \tilde{\mathbf{n}} \rangle_l \langle \tilde{\mathbf{n}} \rangle_l)^2 \right\rangle_l}{1 - \left(\left\langle \tilde{\mathbf{n}}^2 \right\rangle_l - \langle \tilde{\mathbf{n}} \rangle_l^2\right)}. \end{aligned} \tag{19}$$

It seems that the first and second terms in the numerator will lead to second-order terms in the expression for energy. However, it is not so. Using (13) and (16), one has

$$\left\langle \left(\tilde{\bar{\mathbf{n}}} \cdot \frac{\langle \bar{\mathbf{n}} \rangle}{\|\langle \bar{\mathbf{n}} \rangle\|}\right)^2 \right\rangle_l = \frac{\left\langle \left(-\frac{1}{2}(\tilde{\mathbf{n}}^2 - \langle \tilde{\mathbf{n}} \rangle_l^2) + \tilde{\mathbf{n}}\langle \tilde{\mathbf{n}} \rangle_l - \langle \tilde{\mathbf{n}} \rangle_l \langle \tilde{\mathbf{n}} \rangle_l\right)^2 \right\rangle_l}{1 - \left(\left\langle \tilde{\mathbf{n}}^2 \right\rangle_l - \langle \tilde{\mathbf{n}} \rangle_l^2\right)}. \tag{20}$$

One can see that this term has a fourth-order dependence with respect to perturbations. Thus, one can neglect this term in the expression for energy. Consequently, one has

$$E(\mathbf{X}, t) \approx E_{\min} + (E_{\max} - E_{\min})\left(\left\langle \tilde{\mathbf{n}}^2 \right\rangle_l - \langle \tilde{\mathbf{n}} \rangle_l^2\right). \tag{21}$$

Now assume that the acoustic field is periodic in time. The average energy over the period of the acoustic field T is equal to

$$\overline{E}(X) \approx E_{\min} + (E_{\max} - E_{\min})\left(\left\langle\langle\tilde{\mathbf{n}}^2\rangle_l\right\rangle_T - \left\langle\langle\tilde{\mathbf{n}}\rangle_l^2\right\rangle_T\right). \tag{22}$$

It is physically obvious that both terms in (22) are proportional to the intensity of sound, and thus, if the intensity of sound depends on coordinates, some average force acting on the particle will appear.

One can express this force as the negative of the gradient of the energy. Thus, one has

$$\mathbf{F}(\mathbf{X}) = -(E_{\max} - E_{\min})\nabla_x\left(\left\langle\langle\tilde{\mathbf{n}}^2\rangle_l\right\rangle_T - \left\langle\langle\tilde{\mathbf{n}}\rangle_l^2\right\rangle_T\right). \tag{23}$$

The appearance of this average force pushing the particle out from the stronger acoustic field in the liquid crystal is referred to as the "acoustic wind." In accordance with (26), a higher gradient of intensity will provide a bigger force acting on the particles. It is possible to use an acoustic field with high frequencies of "slow," highly attenuating, waves of the viscous type. The major interest, due to short wavelength at low frequencies is the propagation of the so-called *fast shear wave* in the LC, which is an analogue to viscous waves in regular liquids. This wave can be easily generated within the LC simply by the in-plane vibration of the substrate.

3. PERIODIC SHEAR FIELD OF THE FAST SHEAR WAVE

The governing equations for the fast shear wave in the nematic LC can be presented as [7]

$$\partial_i \nu_i = 0, \tag{24}$$

$$\rho\frac{d\nu_i}{dt} - \partial_k\sigma'_{ik} + \partial_i\tilde{p} = 0, \tag{25}$$

$$\frac{d\tilde{n}_i}{dt} = \Omega_{ik}n_k + \lambda(\delta_{ik} - n_i n_l)n_k\nu_{kl}, \tag{26}$$

where the tensor of viscous stress for an incompressible nematic is

$$\sigma'_{ik} = 2\eta_l\nu_{ik} + (\eta_3 - 2\eta_1)(n_i n_l \nu_{kl} + n_k n_l \nu_{il}) + (\bar{\eta}_2 + \eta_1 - 2\eta_3)n_i n_k n_l n_n \nu_{lm} \tag{27}$$

and

$$\bar{\eta}_2 = \eta_2 + \eta_5 - 2\eta_4. \tag{28}$$

We denote the non-perturbed director of the LC as $n_i = (\sin\Theta, 0, \cos\Theta)$ and $\tilde{\mathbf{n}}_i$ as the perturbation of the director in the wave. Thus, only three combined independent viscosity coefficients are significant in this case.

Here we denote the tensor of the velocity of deformation as

$$\nu_{ik} = \frac{1}{2}(\partial_i\nu_k + \partial_k\nu_i) = \frac{1}{2}\begin{pmatrix} 2\partial_x\nu_x & (\partial_x\nu_y + \partial_y\nu_x) & (\partial_x\nu_z + \partial_z\nu_x) \\ (\partial_y\nu_x + \partial_x\nu_y) & 2\partial_y\nu_y & (\partial_y\nu_z + \partial_z\nu_y) \\ (\partial_z\nu_x + \partial_x\nu_z) & (\partial_z\nu_y + \partial_y\nu_z) & 2\partial_z\nu_z \end{pmatrix}, \tag{29}$$

and the tensor of rotation of LC as

$$\Omega_{ik} = \frac{1}{2}(\partial_i\nu_k - \partial_k\nu_i) = \frac{1}{2}\begin{pmatrix} 0 & (\partial_x\nu_y - \partial_y\nu_x) & (\partial_x\nu_z - \partial_z\nu_x) \\ (\partial_y\nu_x - \partial_x\nu_y) & 0 & (\partial_y\nu_z - \partial_z\nu_y) \\ (\partial_z\nu_x - \partial_x\nu_z) & (\partial_z\nu_y - \partial_y\nu_z) & 0 \end{pmatrix}. \tag{30}$$

Equation (24) represents the condition of incompressibility of LC, Eq. (25) is the dynamic equation for LC flow, and at last, Eq. (26) defines the dynamic of the director field induced by the LC flow.

The first term in the right-hand part of Eq. (26) represents simply the rotation of the director field, related to the rotation of a liquid particle. The second term represents the kinetic effect of the LC building blocks' relaxation to the "equilibrium" direction (one can find a detailed discussion of LC hydrodynamic and stationary drag of spherical particles in the paper by H. Stark [8]). We will suppose that this term is always small for the frequency range (What is the frequency range?) being considered, and thus, the liquid crystal is always aligned in accordance with the local movement of the fluid.

It means that we will put in (26) $\lambda = 0$ and, thus, will suppose that

$$\frac{d\tilde{n}_i}{dt} = \Omega_{ik} n_k \tag{31}$$

Equation (24) can be satisfied as usual by the field of the velocity, which is the circulation of some vector field Ψ

$$\nu = \nabla \times \Psi. \tag{32}$$

Considering the field generated within the fluid by the homogeneous shear movement of the boundary surface, one has

$$\begin{pmatrix} \nu_x \\ \nu_y \\ \nu_z \end{pmatrix} = \begin{vmatrix} \mathbf{i} & \mathbf{j} & \mathbf{k} \\ \partial_x & \partial_y & \partial_z \\ \Psi_x & \Psi_y & \Psi_z \end{vmatrix} = \begin{pmatrix} -\partial_z \Psi_y \\ \partial_z \Psi_x \\ 0 \end{pmatrix}. \tag{33}$$

We take into account here that the field does not depend on the in-plane coordinates (x, y). One has

$$\begin{pmatrix} -\partial_z \Psi_y \\ \partial_z \Psi_x \\ 0 \end{pmatrix} = \begin{pmatrix} -\left(\frac{1}{2}\eta_3 + \sin^2\Theta\cos^2\Theta(\bar{\eta}_2 + \eta_1 - 2\eta_3)\right)\partial_z^3 \Psi_y \\ \left(\eta_1 + \frac{1}{2}(\eta_3 - 2\eta_1)\cos^2\Theta\right)\partial_z^3 \Psi_x \\ -((\eta_3 - 2\eta_1) + (\bar{\eta}_2 + \eta_1 - 2\eta_3)\cos^2\Theta)\sin\Theta\cos\Theta\partial_z^3 \Psi_y + \partial_z \tilde{p} \end{pmatrix}, \tag{34}$$

or

$$\tilde{p} = -((\eta_3 - 2\eta_1) + (\bar{\eta}_2 + \eta_1 - 2\eta_3)\cos^2\Theta)\sin\Theta\cos\Theta\partial_z \nu_x, \tag{35}$$

$$\dot{\nu}_x = \mu_{\|}\partial_z^2 \nu_x, \tag{36}$$

$$\dot{\nu}_y = \mu_{\perp}\partial_z^2 \nu_y, \tag{37}$$

where we introduced effective "kinetic viscosity coefficients" for two types of wave polarization

$$\mu_{\|} = \frac{1}{\rho}\left(\frac{1}{2}\eta_3 + \sin^2\Theta\cos^2\Theta(\bar{\eta}_2 + \eta_1 - 2\eta_3)\right) \tag{38}$$

$$\mu_{\perp} = \frac{1}{\rho}\left(\eta_1 + \frac{1}{2}(\eta_3 - 2\eta_1)\cos^2\Theta\right). \tag{39}$$

One can see that the waves with two polarizations (in plane of the orientation of the director and in orthogonal direction) are propagating separately and represent simple viscous waves. For orthotropic orientation of the nematic $\Theta = \frac{\pi}{2}$, the viscosity coefficients (42) and (43) are obviously equal. Thus, one immediately comes to the solution for the velocity of the periodic field characterized by a frequency ω inside the LC

$$\nu_{\|,\perp} = \nu_{0\|,\perp} e^{-\frac{\omega}{C_{\|,\perp}}(1+i)z}, \tag{40}$$

where we introduced the "velocity" of the viscous wave

$$C_{\|,\perp} = \sqrt{2\omega\mu_{\|,\perp}}. \tag{41}$$

One can now find the variation of the director field in the frequency domain for in-plane polarization)

$$\tilde{n}_{x\omega} = -\frac{\nu_{0\|}}{2C_{\|}}(i-1)e^{\frac{\omega}{C_{\|}}(1+i)z}\cos\Theta; \quad \tilde{n}_{z\omega} = -\frac{\nu_{0\|}}{2C_{\|}}(i-1)e^{\frac{\omega}{C_{\|}}(1+i)z}\sin\Theta \tag{42}$$

and

$$\tilde{n}_{y\omega} = -\frac{\nu_{0\|}}{2C_{\perp}}(i-1)e^{-\frac{\omega}{C_{\perp}}(1+i)z}\cos\Theta, \tag{43}$$

for orthogonal polarization.

Solutions (40) and (42) are formally valid under the following condition:

$$\frac{\nu}{V} \ll 1. \tag{44}$$

The characteristic viscous wave "velocity" for "water" (kinematic viscosity $\mu{\sim}10^{-2}$ cm/sec) and frequency 10 Hz is equal approximately to 1.12 cm/sec. Thus, for all reasonable frequencies of interest and reasonable intensities of the sound condition, (44) can be easily satisfied.

However, one remark about the obtained solution must be stated. One cannot satisfy arbitrary boundary conditions for the director field on the surface of the LC only with the help of the above solution. To do this correctly, one must also take into account the existence of the second (slow) shear wave, which can provide the conjugation between the obtained solution and real boundary conditions. The "slow" wave attenuates significantly faster than the "fast wave." This means that a thin transition layer near the wall will appear. The thickness of this transition layer will be in the range of the slow wavelength. However, instead of focusing on this detail, we will focus on the major effects of the acoustic wind on particle assembly and self-assembly.

The real parts of expressions (40), (42), and (43) represent the amplitude of the physical field of the director perturbations.

One has

$$\mathrm{Re}\tilde{n}_{x\omega} = \frac{\nu_{0||}}{2C_{||}} e^{\frac{\omega}{C_{||}}z} \left(\cos\frac{\omega}{C_{||}} z - \sin\frac{1}{\sqrt{2}}\frac{\omega}{C_{||}} z \right) \cos\Theta; \tag{45}$$

$$\mathrm{Re}\tilde{n}_{z\omega} = \frac{\nu_{0||}}{2C_{||}} e^{\frac{\omega}{C_{||}}z} \left(\cos\frac{\omega}{C_{||}} z - \sin\frac{1}{\sqrt{2}}\frac{\omega}{C_{||}} z \right) \sin\Theta; \tag{46}$$

$$\mathrm{Re}\tilde{n}_{y\omega} = \frac{\nu_{0\perp}}{2C_{\perp}} e^{-\frac{\omega}{C_{||}}z} \left(\cos\frac{\omega}{C_{\perp}} z - \sin\frac{1}{\sqrt{2}}\frac{\omega}{C_{\perp}} z \right) \cos\Theta. \tag{47}$$

4. ACOUSTIC SHEAR WIND IN THE NEMATIC LC

To calculate the force acting on a particle embedded in a nematic LC in the presence of the fast shear wave, one must combine the results presented in previous two parts of article, i.e., to calculate the force (26) with the help of expressions (45)–(47).

Let us now express the director field in terms of the position of the center mass of the particle (0,0, Z) and coordinate along particle l.

One has

$$z = Z + lN_z = Z + l\sqrt{1 - (N_x^2 + N_y^2)}. \tag{48}$$

Here we excluded N_z, taking into account that

$$N_x^2 + N_y^2 + N_x^2 = 1. \tag{49}$$

Taking into account that in our approximation the particle is oriented along the average director, one can replace the components of **N** in (48) and (49) with associated approximate expressions

$$N_x \approx n_{0x} + \langle\tilde{n}_x\rangle_l = \sin\Theta + \langle\tilde{n}_x\rangle_l \quad N_y \approx \langle\tilde{n}_y\rangle_i. \tag{50}$$

Formally, one has

$$z = Z + l\sqrt{\cos^2\Theta - (2\langle\tilde{n}_x\rangle_l + \langle\tilde{n}_x\rangle_l^2 + \langle\tilde{n}_y\rangle_l^2)}. \tag{51}$$

However, in the acoustic approximation, for larger tilt angles: $\cos^2\Theta \gg \langle\tilde{n}_x\rangle_l$ one can neglect the second term under the square root in (51). Thus, the expressions for the director perturbations can be written as

$$\tilde{n}_{x\omega} = \frac{\nu_{x0}}{2C_{\|}}(1-i)\cos\Theta(1-\lambda\cos 2\Theta)e^{-\frac{\omega Z}{C_{\|}}}\mathrm{e}^{i\omega\left(t-\frac{Z}{C_{\|}}\right)}e^{-\frac{\omega}{C_{\|}}(1+i)l\cos\Theta}, \tag{52}$$

$$\tilde{n}_{z\omega} = -\frac{\nu_{x0}}{2C_{\|}}(1-i)\sin\Theta(1-\lambda\cos 2\Theta)e^{-\frac{\omega Z}{C_{\|}}}\mathrm{e}^{i\omega\left(t-\frac{Z}{C_{\|}}\right)}e^{-\frac{\omega}{C_{\|}}(1+i)l\cos\Theta}, \tag{53}$$

$$\tilde{n}_{y\omega} = \frac{\nu_{y0}}{2C_{\|}}(1-i)\cos\Theta(1-\lambda)e^{-\frac{\omega Z}{C_{\perp}}}\mathrm{e}^{i\omega\left(t-\frac{Z}{C_{\perp}}\right)}e^{-\frac{\omega}{C_{\perp}}(1+i)l\cos\Theta}. \tag{54}$$

Here we explicitly introduce the dependence of the director field over time.

Now, one has to calculate the value $Q = (\langle\langle\tilde{\mathbf{n}}^2\rangle_l\rangle_T - \langle\langle\tilde{\mathbf{n}}\rangle_l^2\rangle_T)$. One has

$$Q = (\langle\langle\tilde{\mathbf{n}}^2\rangle_l\rangle_T - \langle\langle\tilde{\mathbf{n}}\rangle_l^2\rangle_T) = (Q_x + Q_z) + Q_y. \tag{55}$$

Here,

$$\begin{aligned} Q_x = {} & \left(\frac{\nu_{x0}}{\omega L}\right)^2(1-\lambda\cos 2\Theta)^2\left(\frac{\omega}{2C_{\|}}\cos\Theta\sinh\left(\frac{\omega L}{C_{\|}}\cos\Theta\right)\right. \\ & \left. - \begin{pmatrix} \cosh^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\cdot\sin^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right) + \\ + \sinh^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\cdot\cos^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\end{pmatrix}\right)e^{\frac{2\omega Z}{C_{\|}}}, \end{aligned} \tag{56}$$

$$\begin{aligned} Q_z = {} & \left(\frac{\nu_{x0}}{\omega L}\right)^2(1-\lambda\cos 2\Theta)^2\frac{\sin^2\Theta}{\cos^2\Theta}\left(\frac{\omega}{2C_{\|}}\cos\Theta\sinh\left(\frac{\omega L}{C_{\|}}\cos\Theta\right)\right. \\ & \left. - \begin{pmatrix} \cosh^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\cdot\sin^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right) + \\ + \sinh^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\cdot\cos^2\left(\frac{\omega L}{2C_{\|}}\cos\Theta\right)\end{pmatrix}\right)e^{\frac{2\omega Z}{C_{\|}}}, \end{aligned} \tag{57}$$

$$\begin{aligned} Q_y = {} & \left(\frac{\nu_{y0}}{\omega L}\right)^2(1-\lambda)^2\left(\frac{\omega}{2C_{\perp}}\cos\Theta\sinh\left(\frac{\omega L}{C_{\perp}}\cos\Theta\right)\right. \\ & \left. - \begin{pmatrix} \cosh^2\left(\frac{\omega L}{2C_{\perp}}\cos\Theta\right)\cdot\sin^2\left(\frac{\omega L}{2C_{\perp}}\cos\Theta\right) + \\ + \sinh^2\left(\frac{\omega L}{2C_{\perp}}\cos\Theta\right)\cdot\cos^2\left(\frac{\omega L}{2C_{\perp}}\cos\Theta\right)\end{pmatrix}\right)e^{\frac{2\omega Z}{C_{\perp}}}. \end{aligned} \tag{58}$$

It follows from (56) and (57), and in accordance with (233), that force acting on the particle is proportional to $\frac{d}{dz}(Q_x + Q_y + Q_z)$ and acts in z-direction.

The general expression is very long. However, it can be drastically simplified if one suggests that the size of a particle is significantly smaller than the wavelength and, thus,

$$\frac{\omega}{2C_{||,\perp}} L \cos\Theta \ll 1. \tag{59}$$

Retaining only the major terms in the final expression with respect to $\frac{\omega}{2C_{||,\perp}} L \cos\Theta$, one has

$$Q_x = \frac{\nu_{x0}^2 L^2}{48 \cdot \mu_{||}^2} (1 - \lambda \cos 2\Theta)^2 \cos^4\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{||}}}}, \tag{60}$$

$$Q_z = \frac{\nu_{x0}^2 L^2}{48 \cdot \mu_{||}^2} (1 - \lambda \cos 2\Theta)^2 \cdot \sin^2\Theta \cdot \cos^2\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{||}}}}, \tag{61}$$

$$Q_y = \frac{\nu_{y0}^2 L^2}{48 \cdot \mu_{\perp}^2} (1 - \lambda)^2 \cos^4\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{\perp}}}}. \tag{62}$$

One can see that it is possible to separate effect of excitation in plane of tilt and for orthogonal excitation. One has

$$Q_{||} = Q_x + Q_y = \frac{\nu_{x0}^2 L^2}{48 \cdot \mu_{||}^2} (1 - \lambda \cos 2\Theta)^2 \cos^2\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{||}}}}, \tag{63}$$

$$Q_{\perp} = Q_y = \frac{\nu_{y0}^2 L^2}{48 \cdot \mu_{\perp}^2} (1 - \lambda)^2 \cos^4\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{\perp}}}}. \tag{64}$$

Appropriate forces are

$$F_{z||} = \frac{\nu_{x0}^2 L^2 (E_{\max} - E_{\min})}{48 \cdot \mu_{||}^2} \sqrt{\frac{2\omega}{\mu_{||}}} (1 - \lambda \cos 2\Theta)^2 \cos^2\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{||}}}}, \tag{65}$$

$$F_{z\perp} = \frac{\nu_{y0}^2 L^2 (E_{\max} - E_{\min})}{48 \cdot \mu_{\perp}^2} \sqrt{\frac{2\omega}{\mu_{\perp}}} (1 - \lambda)^2 \cos^4\Theta \cdot \mathrm{e}^{-Z\sqrt{\frac{2\omega}{\mu_{\perp}}}}. \tag{66}$$

Three observations can be made from this result. The first is that the maximal force is reaching if Θ=0. The second observation is that there exists characteristic length, which defines the maximal force acting on particle. This length is

$$L_\nu = \frac{48\mu^2_{||,\perp}\lambda_{||,\perp}}{\nu^2_{x,y0}L^2}; \quad \lambda_{||,\perp} = \sqrt{\frac{\mu_{||,\perp}}{2\omega}}. \tag{67}$$

The last observation is related with effect of polarization of the exited waves in respect to director of tilt plane. Taking the ratio between forces generated by waves with different polarizations, one has

$$R = \frac{\nu^2_{x0}}{\nu^2_{y0}}\left(\frac{\mu_\perp}{\mu_{||}}\right)^{5/2}\frac{(1-\lambda\cos\Theta)^2}{(1-\lambda)^2\cos^2\Theta}\cdot e^{-Z\sqrt{\frac{2\omega}{\mu_{||}}}\left(1-\sqrt{\frac{\mu_{||}}{\mu_\perp}}\right)}. \tag{68}$$

One can recognize in (68) effects of three different natures. The first factor is that viscous waves having different polarizations attenuate with distance differently, which is reflecting in the exponential factor. The second factor is geometrical. Waves polarized in the plane of director orientation interact with director differently than the waves with orthogonal polarization. If tilt angle tends to zero, this difference disappears. The last mechanism is related with the fact that in anisotropic materials, shear waves of different polarization have different impedances.

5. SOME ESTIMATES

It is convenient to compare the force that can be reached with the help of acoustic shear wind with the gravity force acting on the particle. Let us consider, as an example, an SWCNT with the diameter 10^{-6} cm and the length 10^{-3} cm, which are typical geometric parameters. We will assume that the LC layer has a thickness of 10^{-2} cm and will determine the maximal force that can be provided at this distance. We will suppose for simplicity that the nonperturbed LC has the orthotropic alignment, and thus, $\cos\Theta=1$. Let us consider an LC with a kinematic viscosity equal to 10^{-2} cm^2/sec, one can determine the optimal frequency must in the range of 10^2 sec^{-1}.

If one estimates the value $(E_{\max} - E_{\min})$ as $\sim(Ka)$, where $K\sim10^{-6}$ *Dyn* (this is the elastic modulus of the liquid crystal), and considering the vibration velocity to be equal to 1 cm/sec, one has for characteristic length

$$L_\nu = \frac{48\mu^2_{||,\perp}\lambda_{||,\perp}}{\nu^2_{x,y0}L^2} = \frac{48\cdot 10^{-4}\,\frac{\text{cm}^4}{\text{sec}}\cdot 10^{-2}\ \text{cm}}{1\,\frac{\text{cm}^2}{\text{sec}^2}\cdot 10^{-6}\ \text{cm}^2} = 48\ \text{cm}, \tag{69}$$

and the force is

$$F = \frac{10^{-6} Dyn \cdot 10^{-3}\ \text{cm}}{48\ \text{cm}} = 2.8 \cdot 10^{-11} Dyn. \tag{70}$$

It is obvious that, at shorter distances, the force will increase exponentially. In the meantime, the mass of considering nanotube is equal to approximately $3.14 \cdot 10^{-7}$ G, and, thus, the gravity force acting on the particle is equal to

$$F = M \cdot g = 3.14 \cdot 10^{-17} G \cdot 9.81 \cdot 10^{3}\,\text{cm}/\text{sec}^{2} \approx 3.10^{-13} Dyn. \tag{71}$$

Thus, the acoustic shear wind force is practically two orders of magnitude larger than the gravity force, and the sedimentation time can be decreased to the order of minutes. This compares favorably with the hydrodynamic method of sedimentation while providing a more homogenous assembly. Frequency of interest can be estimated from the condition that viscous wavelength must be on the order of the layer thickness. This gives the estimate: $\omega \approx 50\,\text{sec}^{-1}$, or $f \approx 7$–$8\,\text{Hz}$.

It is important to mention that ponderomotive forces are universal. As discussed above, the physical nature of the specific force considered here is related with the storage of energy in the form of the energy of deformation of the director field of the LC. From this point of view, it is interesting to also compare this force, specific for LCs, with the ponderomotive force acting on the particle in regular liquids having no long range order. In this case, the energy can be stored only in the form of the average kinetic energy of the particle. The ponderomotive force acting on the particle is equal in this case to

$$F_p = -\nabla (K_T + K_\Omega) \approx -\frac{(K_T + K_\Omega)}{\lambda}, \tag{72}$$

where K_T is the translational part of kinetic energy:

$$K_T = \frac{M \langle \mathbf{V}^2 \rangle_T}{2}, \tag{73}$$

and K_Ω is the rotational energy:

$$K_\Omega = \frac{\langle \hat{\mathbf{J}} \cdot \Omega\Omega \rangle_T}{2}. \tag{74}$$

Here M is the mass of the particle; $\hat{\mathbf{J}}$ is the tensor of particle inertia; $\mathbf{V}$ is the velocity of the particle center of mass; Ω is the angular velocity of particle rotation; and λ is the characteristic length of changing the intensity of acting wave. For a viscous wave,

$$\frac{1}{\lambda} \sim \sqrt{\frac{2\omega}{\mu_{||}}}, \tag{75}$$

assuming (as we did before) that values must be averaged over the period of acting force. We focus here only on the effect of the viscous wave in a regular viscous fluid. The possibility of forces acting on particles from the "regular" acoustic field at very high frequencies with wavelengths of the order of the thickness of the LC layer, will be the subject of future efforts.

In a viscous wave, which can be generated in liquid layer by appropriate vibration of the substrate, the velocity of particle center of mass cannot exceed the velocity of the fluid and thus the velocity of vibration. Considering, as we did above, this velocity equal to 1 cm/sec, one has to kinetic energy of the particulate:

$$K_T = 0.5 * 3.14 \cdot 10^{-17} * (1\,\mathrm{cm/sec})^2 = 1.57 \cdot 10^{-17} Erg. \tag{76}$$

Suggesting that the characteristic wavelength is of the order of thickness of liquid layer $\sim 10^{-2}$ cm, one has an estimate for the ponderomotive force related with gradient of the translational part of the kinetic energy of particle:

$$|F_T| \approx \frac{1.57 \cdot 10^{-17} Erg}{0.01\,\mathrm{cm}} = 1.57 \cdot 10^{-15} Dyn, \tag{77}$$

which is two orders smaller than the gravity force and four orders smaller than the ponderomotive force related with the gradient of the energy of LC deformation. Similarly, one can estimate the rotational part of kinetic energy.

Taking into account that the inertia of rigid rod is equal to $J = \frac{1}{3} ML^2$, one has

$$K_\Omega = \frac{1}{2} \cdot \frac{1}{3} \cdot 3.14 \cdot 10^{-17} G \cdot (10^{-2}\mathrm{cm})^2 \cdot \left(7 - 8\frac{1}{\mathrm{sec}}\right)^2 = 3.35 \cdot 10^{-20} Erg. \tag{78}$$

Thus, the associated ponderomotive force is equal to

$$F_\Omega = 3.35 \cdot 10^{-18} Dyn. \tag{79}$$

This is five orders smaller than gravity force and seven orders smaller than the above-considered ponderomotive force acting on the particle in LCs.

Thus, one can see that the effect of liquid crystalline long rage order on the creation of force acting on the particles in shear wave is decisive.

6. CONCLUSIONS

We have considered the average force acting on a particle embedded in liquid crystalline material subjected to an acoustic wave. Our results have shown that, under reasonable conditions, this force acting on nanotubes having a length of ten micrometers can generate forces that are two or more orders higher than the gravity force.

The "shear wind" force is sensitive to the linear size of the particle, and thus, the shear acoustic wind can be used for separation of nanotubes of different lengths. This interaction could also be used for the measurements of the LC viscosity using nanotubes of known length. It is also possible to use this effect to prevent sedimentation of the large particles on the vibrating wall. This in turn can be used for particle separation over ranges of mass and size (for example, to use this effect in combination with electrophoreses).

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